

# Electron spin coherence in semiconductors: Considerations for a spin-based solid state quantum computer architecture

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We theoretically consider coherence times for spins in two quantum computer architectures, where the qubit is the spin of an electron bound to a P donor impurity in Si or within a GaAs quantum dot. We show that low temperature decoherence is dominated by spin-spin interactions, through spectral diffusion and dipolar flip-flop mechanisms. These contributions lead to  $1 - 100 \mu\text{s}$  calculated spin coherence times for a wide range of parameters, much higher than former estimates based on  $T_2^*$  measurements.

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Solid state quantum computation based on the intrinsic two level dynamics of electron spin in semiconductors has attracted widespread attention because the enormous resources of conventional electronics can in principle be applied to develop a scalable quantum computer (QC). In this context, solid state spins in an applied magnetic field are attractive qubit candidates since they comprise perfect two-level systems with potentially long coherence times. Nuclear spins of phosphorus impurities in silicon are potential qubits because they are well isolated from other degrees of freedom [1]. But for such a quantum computer to work, precise electronic control of single nuclear spins must be achieved, a rather daunting task. Electron spins are much easier to control. In that respect, electrons bound to phosphorus impurities in silicon [2] and gallium arsenide quantum dots [3] are promising qubit candidates. However, for quantum computation to be fault-tolerant these electron spins must be coherent for at least  $10^4$  elementary quantum operations [4], which imposes a severe constraint since very long spin coherence times would be needed. In this letter we address the important question of principle involving the fundamental upper bound on electron spin coherence time in proposed semiconductor QC architectures. Since such a device can only work at low temperatures ( $k_B T \ll E_Z$ , where  $E_Z$  is the Zeeman splitting of the spins in an applied magnetic field  $B$ ), we show that the dominant decoherence contribution comes from the unavoidable spin-spin interactions with nuclei and other electrons. This leads to an unsurmountable upper bound on spin coherence, but our calculations indicate that the fault-tolerance criterion can still be satisfied for a wide range of parameters (e.g. quantum dot Fock-Darwin radius  $l_0$ , concentration of  $^{29}\text{Si}$  isotopes  $f$ , etc) defining the QC architecture.

An essential property of electrons in Si:P and GaAs is that their electron spin resonance (ESR) line is inhomogeneously broadened by the hyperfine interaction with nuclear spins [5,6]. This effect leads to a drastic difference between the precessing magnetization of an ensemble of

spins as compared to a single spin or a group of them with the same Larmor frequency. The former decays in a time scale  $T_2^*$  which is dominated by the dephasing effect of the inhomogeneous field distribution. The latter magnetization decays in a time  $T_2$  which is usually many orders of magnitude longer [7]. To extract the single spin coherence time  $T_2$  from an ESR experiment one has to perform a  $\pi/2 - \pi$  spin echo sequence [8], that forces the spins to refocus eliminating the dephasing effect. This leads to a convenient definition for spin coherence: Simply the time it takes for a spin echo envelope to decay to  $1/e$  of its initial value. Since spin echoes usually decay in a quite different fashion from the simple exponential predicted by the Bloch equations, we denote its decay time by  $T_M$  using a notation consistent with existing literature [9]. Spin echo experiments have been performed in dilute Si:P [10,11], but never in GaAs. For the latter material, claims of “long coherence times” have been based on  $T_2^*$  measurements only [12], which do not reveal the ultimate limit on spin coherence set by  $T_M$ , that can in principle be many orders of magnitude longer than  $T_2^*$ . Certainly inhomogeneous broadening imposes severe tuning constraints on one-qubit gates. But fortunately one can still have an universal QC built only with two-qubit gates [13]. Our former work shows that the spread of Zeeman frequencies will only weakly affect two-qubit gates, since the exchange interaction is fairly insensitive to inhomogeneous fields [14]. Therefore we emphasize that the relevant spin coherence time for QC architectures is, in fact,  $T_M$  (not  $T_2^*$ ), and our theoretical finding of truly large values of  $T_M$  ( $\gtrsim \mu\text{s}$ ) compared with the measured  $T_2^*$  ( $\sim \text{ns}$ ) values is quite significant.

Due to extreme sensitivity requirements, ESR in GaAs is usually measured indirectly, for example from the changes in magneto-resistivity in a 2DEG [15] or from photoluminescence [16]. In these experiments one probes ensemble spin properties of moving electrons or recombination pairs respectively, meaning that one should be careful in extrapolating those results to a single localized

spin in a quantum computer environment. Alternative methods to ESR include Faraday rotation [12], but again to study single spin coherence it would be necessary to perform an echo sequence, that is yet to be done using optical methods. Measuring single spin coherence time in semiconductors is a dauntingly difficult task, and therefore the need for theoretical estimates of  $T_M$  becomes acutely necessary. Here we present the first realistic calculation of  $T_M$  for a GaAs quantum dot (QD) based QC architecture (and also for Si:P QC architecture). Our values exceed former coherence estimates based on  $T_2^*$  by three orders of magnitude, and establish quite definitively that fault tolerant quantum computation should be possible in semiconductor QC at low temperatures.

First we discuss the spin-flip rate  $T_1^{-1}$  with corresponding energy transfer to the lattice. Any spin-flip process contributes to exponential decay of the spin echo signal. But bound electrons at low temperatures quite generally have extremely long  $T_1$  ( $\gg T_M$ ), since for the spin to flip, a virtual transition to an excited orbital state must take place, quite different from the conduction electron case where any momentum relaxation event may flip the spin [17] through spin-orbit coupling. The direct phonon emission rate for bound-electrons becomes  $T_1^{-1} \propto [n(E_Z) + 1]B^5$ , where  $n(E_Z) = [\exp(E_Z/k_B T) - 1]^{-1}$  is the Bose occupation number for the emitted phonon wave-vector. Feher and Gere measured  $T_1^{-1} \approx 4 \times 10^{-4} \text{ s}^{-1}$  for Si:P at  $B = 0.32 \text{ T}$  and  $T = 1.25 \text{ K}$  [18]. For GaAs dots with Fock-Darwin radius  $l_0 = 30 \text{ nm}$  and  $B=1 \text{ T}$  one obtains  $T_1^{-1} \approx 200 \text{ s}^{-1}$  [19]. These spin-flip rates when compared to other decoherence mechanisms considered below give negligible contributions, with the possible exception of very large dots ( $l_0 > 100 \text{ nm}$ ), due to the fact that  $T_1^{-1} \propto l_0^8$ , a case we do not consider here since reasonable semiconductor QC architectures are limited by an inter-dot distance of  $50 \text{ nm}$ , necessary for exchange gate operations to work [3].

We now describe the spin decoherence mechanisms of importance to QC architectures. Two electron spins may flip-flop due to their dipolar interaction. But this event is limited by inhomogeneous broadening, because only spins with the same Zeeman splitting can satisfy energy conservation in this process [20]. The rate is given by [9]

$$\frac{1}{T_f} \approx \pi f(\omega_0) \times \frac{\langle \Delta\omega^2 \rangle}{9} \simeq 0.33 \frac{\langle \Delta\omega^2 \rangle}{(\Delta\omega)_I}. \quad (1)$$

Here  $f(\omega_0)$  is the inhomogeneously broadened Gaussian line-shape, which plays the role of the density of states per unit frequency,  $(\Delta\omega)_I$  the inhomogeneous line-width, while  $\langle \Delta\omega^2 \rangle/9$  is the flip-flop contribution to the second moment [8], which is of the order of the transition matrix element squared. It is important to mention that the qubit-qubit dipolar interaction can in principle be included in the Hamiltonian responsible for the quantum algorithm [21], and also may be eliminated using magnetic resonance techniques [8]. But here we have two

reasons to include it in our calculation: First we do not expect forthcoming  $T_M$  measurements to be free from these effects. Second, since correcting dipolar coupling implies additional overhead in QC design, it is interesting to access the amount of error involved by ignoring its presence. Another spin-spin mechanism is spectral diffusion, that happens when the electron spins that generate the echo pulse are subject to fluctuating dipolar or hyperfine fields generated by nuclear spins. A stochastic theory of this effect can be formulated by treating the electron spin Larmor frequency  $\omega$  as a random variable, and calculating the echo envelope amplitude  $M(2\tau)$  using an ensemble average [22]. The result is

$$M(2\tau) \approx M(0) \exp [-(R\delta_A^2/12)(2\tau)^3], \quad (2)$$

where  $\tau$  is the time interval between the  $\pi/2$  and  $\pi$  pulses, and  $R$  the local field relaxation to a Gaussian probability distribution with width  $\delta_A$ . Hence the spectral diffusion (SD) rate is given by  $T_{SD}^{-1} = (R\delta_A^2/12)^{1/3}$ . The decoherence rate follows from all the above contributions by solving the cubic equation for  $T_M^{-1}$ :  $(T_1^{-1} + T_f^{-1})T_M + (T_{SD}^{-1}T_M)^3 = 1$ , and often one finds it to be dominated by a single mechanism. (We give our calculated values for  $T_M$  in Si:P and GaAs quantum dot system in Table 1, where the dominant decoherence mechanism in each case is also listed.)

Now we estimate  $T_M$  for Si:P. For the dipolar mechanism, assuming a qubit separation  $d = 10 \text{ nm}$  we get  $T_f^{-1} = 1 \times 10^3 \text{ s}^{-1}$  and  $2 \times 10^3 \text{ s}^{-1}$  when the qubits are arranged in 1D and 2D lattices respectively. This is the case for  $^{29}\text{Si}$  natural abundance ( $f = 4.67\%$ ) that leads to an experimental line-width of  $2.5 \text{ G}$  [5]. However, for isotopically pure  $^{28}\text{Si}$  (which has zero spin), we expect no inhomogeneous broadening [ $(\Delta\omega)_I \sim \sqrt{\langle \Delta\omega^2 \rangle}$  in Eq. (1)]. In that case,  $T_f^{-1}$  will be much higher:  $T_f^{-1} = (2\langle \Delta\omega^2 \rangle/\pi)^{1/2} = 3 \times 10^5 \text{ s}^{-1}$ ,  $4 \times 10^5 \text{ s}^{-1}$ , dominated by qubit dipolar coupling. The SD rate was measured for natural silicon by Chiba and Hirai [11]. By assuming that fluctuating dipolar fields of  $^{29}\text{Si}$  nuclear spins caused Gaussian spectral diffusion they formulated a theory to calculate the coefficient  $R\delta_A^2$  in (2), which agreed within order of magnitude with the measured  $T_M$  (note that since their P concentration was low  $T_f^{-1}$  was negligible). Therefore we assume their experimental value of  $T_{SD}^{-1} = 3.3 \times 10^3 \text{ s}^{-1}$  as a reliable estimate for the SD rate for a single P spin in a Si matrix, and by adding the dipolar flip-flop rate we are able to get a phenomenological estimate for the decoherence rate when the P spins are arranged in a quantum computer geometry, as opposed to a dilute randomly doped sample. Hence we get  $T_M^{-1} \approx 4 \times 10^3 \text{ s}^{-1}$  in both 1D and 2D geometries, noting that here both spin-spin mechanisms contribute with the same order of magnitude. An interesting consequence of the interplay of these two mechanisms is that  $T_M^{-1}$  displays a minimum as a function of the  $^{29}\text{Si}$  fraction  $f$ ,

an effect not yet noted in the literature (Fig. 1). The  $T_{SD}^{-1}$  contribution is proportional to  $f^{2/3}$ , since the probability of finding a pair of  $^{29}\text{Si}$  [analogous to  $p_{ij}$  in (10) below] is proportional to  $f^2$ , and then we take a cubic root due to the non-exponential decay in (2). However, the dipolar flip-flop rate is decreased when we increase  $f$ :  $T_f^{-1} \propto (\Delta\omega)^{-1} \propto f^{-1/2}$ . This happens because the  $^{29}\text{Si}$  is the source of hyperfine broadening and the mean square deviation of the hyperfine field is proportional to  $f$ . It would be interesting to fit these results to measurements on isotopically purified samples; to date there is only one measurement, with  $f = 0.12\%$ , in which  $T_M^{-1}$  decreased by a factor of 2 [10] (the P concentration was the same as in Fig. 1,  $c = 4 \times 10^{16} \text{ cm}^{-3}$ ). This is consistent with our result. For the case of a 2D Si:P QC we find that  $T_M^{-1}$  will attain a minimum when  $f \approx 2\%$ , suggesting that natural silicon is a good choice for QC architectures.

Turning to the GaAs-QD, we note that inhomogeneous broadening should be much stronger: We estimate a line-width of at least 50 G due to hyperfine interactions, since all nuclei have spin 3/2 (S impurities in GaAs have 500 G of broadening [6]). This together with the fact that the qubits will be much further apart ( $d = 50 \text{ nm}$ ) leads to  $T_f^{-1} < 10^{-3} \text{ s}^{-1}$  ( $T_f^{-1} \propto d^{-6}$ ). We now calculate the SD rate for these spins. NMR in GaAs reveals a composition of  $^{75}\text{As}$  (50%),  $^{69}\text{Ga}$  (30.2%), and  $^{71}\text{Ga}$  (19.8%) [23]. The homogeneous line-width of the  $^{75}\text{As}$  system (fcc lattice with  $a_0 = 5.65 \text{ \AA}$ ) is  $\delta_B = 5.56 \times 10^3 \text{ s}^{-1}$  [24], indicating that these nuclei are flip-flopping every 300  $\mu\text{s}$ . Since the hyperfine interaction depends on the position of each nucleus, the electron feels a different field if a pair of nuclei is up-down as opposed to down-up (here we will neglect the contribution of nuclei outside the QD wave function; they would produce a dipolar field which shifts the electron frequency by at most  $\delta\omega \sim \gamma_e\gamma_n\hbar/z_0^3 \sim 20 \text{ s}^{-1} \ll \delta_r = T_1^{-1} \sim 200 \text{ s}^{-1}$ , the intrinsic line-width due to QD spin relaxation mentioned above; this is certainly not the case in Si:P, where the nuclei outside the donor wave function give the dominant contribution [11]). To calculate the spectral diffusion rate we consider a three spin Hamiltonian  $H = H_0 + V$ , where

$$H_0 = \hbar\omega_i I_{iz} S_z + \hbar\omega_j I_{jz} S_z, \quad (3)$$

$$V = \hbar b_{ij} (I_{i+} I_{j-} + I_{i-} I_{j+}) - 4\hbar b_{ij} I_{iz} I_{jz}, \quad (4)$$

$$b_{ij} = -\frac{1}{4}\gamma_n^2\hbar \frac{1 - 3\cos^2\theta_{ij}}{R_{ij}^3}, \quad (5)$$

$$\omega_i = \omega_h \exp\left(-\frac{X_i^2 + Y_i^2}{l_0^2}\right) \cos^2\left(\pi \frac{Z_i}{z_0}\right). \quad (6)$$

Here  $H_0$  is the diagonal part of the hyperfine interaction (the off-diagonal component can be neglected since electron-nucleus flip-flop is forbidden by energy conservation), while  $V$  is the dipolar coupling between two nuclei.  $\mathbf{I}_i$  is the spin operator for the nucleus located at

$\mathbf{R}_i = (X_i, Y_i, Z_i)$ ,  $\gamma_n = 4.58 \times 10^3 \text{ (s G)}^{-1}$  the gyromagnetic ratio for  $^{75}\text{As}$ ,  $R_{ij}$  the distance between the two flip-flopping nuclei and  $\theta_{ij}$  the angle between this vector and the magnetic field. The hyperfine frequency is given by

$$\omega_h = \frac{8\pi}{3}\gamma_e\gamma_n\hbar |\Psi(0)|^2 = \frac{16}{3} \frac{\gamma_e\gamma_n\hbar a_0^3}{z_0 l_0^2} d(As), \quad (7)$$

where  $d(As) = |u'(0)|^2$  is the electronic density on the nuclei and  $l_0$  the Fock-Darwin length. The electron frequency change due to a nuclear flip-flop is then  $\Omega_{ij} = \omega_i - \omega_j$ , while the energy changes by  $\hbar\Omega_{ij}/2$  in this process. Since  $|b_{ij}| \lesssim |\Omega_{ij}|$  we can apply perturbation theory, and the rate becomes [11]

$$\Gamma_{ij} = 2\pi b_{ij}^2 g_B(\Omega_{ij}/2), \quad (8)$$

$$g_B(\omega) = \frac{1}{\sqrt{2\pi}\delta_B} \exp\left(-\frac{\omega^2}{2\delta_B^2}\right). \quad (9)$$

The line-width  $\delta_B$  of the nuclear system enters to guarantee energy conservation: the change in the Zeeman energy of the electron is compensated by the spin-spin interaction of the nuclear system. The SD rate can now be estimated by summing over all pairs  $ij$ , as long as they can flip-flop:

$$\frac{1}{12} R \delta_A^2 = \frac{1}{12} \sum_{i < j} \Gamma_{ij} \Omega_{ij}^2 \left[1 - \frac{f_A(\Omega_{ij})}{f_A(0)}\right] p_{ij}(T), \quad (10)$$

$$f_A(\omega) = \frac{1}{\sqrt{2\pi}\delta_r} \exp\left(-\frac{\omega^2}{2\delta_r^2}\right). \quad (11)$$

Assuming the nuclei are in thermal equilibrium we get

$$p_{ij}(T) = \left(1 + \frac{\cosh 3x}{2 \cosh 2x + 3 \cosh x + 2}\right)^{-1}, \quad (12)$$

with  $x = \hbar\gamma_n B/k_B T$ . For  $B = 1 \text{ T}$  and  $T \gg 1 \text{ mK}$  we assume the high  $T$  limit  $p_{ij} \approx 7/8$ . By performing the sum (10) numerically, we estimate  $T_{SD}^{-1} \sim 10^4 \text{ s}^{-1}$ , that shows for QD's with  $l_0 \lesssim 100 \text{ nm}$  spectral diffusion due to hyperfine field fluctuation dominates. This rate depends on B field intensity (Fig. 2), since  $l_0$  decreases when  $B$  is increased, and also varies by about a factor of 2 when the tilting angle with  $z$  direction is changed [see Eq. (5)].

Finally, we wish to comment on the validity of the approximations employed here. The SD decay given by Eq. (2) is only valid for  $R\tau \ll 1$  [22], and also Eq. (10) assumes a generalization of this two parameter model to several parameters  $\Gamma_{ij}$ ,  $\Omega_{ij}$ . Certainly a more rigorous theory of SD due to nuclear spins needs to be developed if one wants to go beyond the order of magnitude estimates given here. In particular, a rigorous theory for the nuclear flip-flop rate [Eq. (8)] is mandatory for a precise description of this phenomena. Our estimated coherence

times, summarized in Table 1, should be compared with the longest gating time in the corresponding QC architecture. For the parameters chosen here, the exchange time will be  $\tau_J \sim \hbar/0.1 \text{ meV} \sim 1 \text{ ps}$ , with a typical exchange coupling [3] of 0.1 meV. A single qubit rotation (Rabi flop) can be done with an ESR field 20 times smaller than the applied field, that leads to a  $\pi/2$  rotation time  $\tau_R \sim 20/\gamma B \sim 0.1 \text{ ns}$ , 0.5 ns at  $B = 1 \text{ T}$  for Si:P, GaAs-QD respectively. These time scales lead to a quality factor  $T_M/\tau_R > 10^4$  for both architectures. Also,  $T_M/\tau_J \sim 10^6 - 10^8$  (since  $\tau_J \sim 1 \text{ ps}$  and  $T_M \sim 1 - 100 \mu\text{s}$ ), which implies a large number ( $\gg 10^4$ ) of coherent gate operations allowing convenient fault tolerant computation well within the currently estimated ( $10^{-4}$ ) error correction scheme. Hence the electron spin in semiconductors is confirmed as a competitive qubit candidate, with the effective low temperature upper bound on the coherence time given by  $1 - 100 \mu\text{s}$  under quite general conditions. The authors acknowledge discussions with J. Fabian, X. Hu, A. Kaminski, B. Koiller, and I. Žutić. This work is supported by ARDA, LPS, US-ONR, and NSF.

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Architecture	$T_M [\mu\text{s}]$	$Q = T_M/\tau_R$	Dominant Mech.
Si:P/natural Si	200	$10^6$	Dip./Spec. Diff.
Si:P/pure $^{28}\text{Si}$	2	$10^4$	Dipolar
GaAs-QD	50	$10^5$	Spec. Diff.

TABLE I. Coherence times  $T_M$ , quality factors, and dominant decoherence mechanisms for three quantum computer architectures. We assume the qubits are disposed in a 2D square lattice of side 10 nm and 50 nm for the case of Si:P and GaAs-QD respectively. For the GaAs-QD architecture, Fock-Darwin radius is assumed 30 nm.

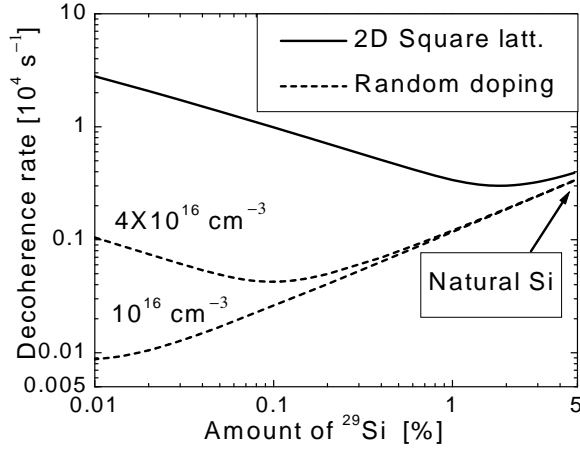


FIG. 1. Depicts the competition between the dipolar flip-flop rate and spectral diffusion for a bound-electron spin in Si:P, leading to a minimum in the decoherence rate as a function of the  $^{29}\text{Si}$  fraction. We show calculations for a 2D quantum computer architecture with qubit separation of 10nm, and for the most common experimental situation of random phosphorus doping, with concentrations  $1 - 4 \times 10^{16} \text{ cm}^{-3}$ .

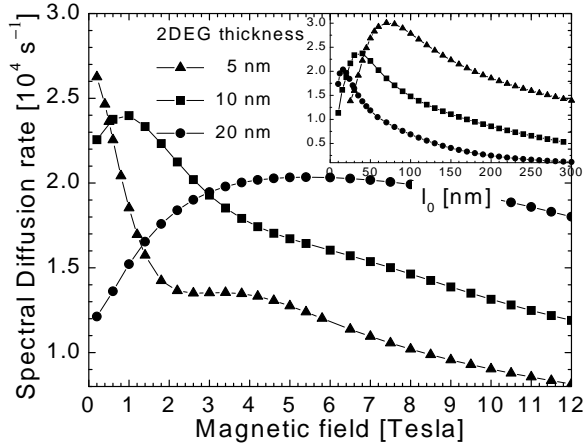


FIG. 2. Calculated spectral diffusion rate as a function of magnetic field and quantum dot Fock-Darwin radius  $l_0$  (inset). For the B field plot the dot transverse confinement length is set to 50 nm. The spectral diffusion mechanism completely dominates the decoherence rate for small dots ( $l_0 < 100 \text{ nm}$ ).